

## Oscillation phenomenon of transition temperatures of coupled magnetic planes

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### Abstract

Oscillatory behavior of transition temperature in super lattice, Ni/Au/Ni, has been observed as a function of spacer layer, Au. The observed oscillation period is almost the half period of interlayer exchange coupling. The high temperature susceptibility of a two - dimensional lattice is evaluated within the Bethe-Peierls-Wiess approximation in the presence of a random field with square distribution. This susceptibility is used to evaluate the transition temperature of coupled planes as a function of spacer thickness. The calculated transition temperature of coupled planes oscillates as half period of interlayer exchange coupling and falls below the results of the uncoupled films at some values of the average spacer thickness that were experimentally observed. Additionally, the transition temperature depends on the distribution function of random field at the small thickness of spacer.

**Keywords:** coupled magnetic planes, transition temperature, random field, interlayer exchange coupling

### 1. Introduction

Magnetic multilayers have provided many interesting physical properties in the field of magnetism, and much attention has been paid to this research area both experimentally and theoretically as it may have potential applications in technology [1-4]. Recently it has been observed that the Curie temperature,  $T_c$ , and ground state moments in exchange coupled magnetic multilayers oscillate with the spacer thickness like the absolute value of interlayer exchange coupling ( $J_{12}$ ) [2,5]. The Curie temperature of a ferromagnetic layer is proportional to the average exchange energy per atom within the mean field theory, and to  $J_{12}$ . In layered magnetic metallic structures, the interlayer exchange coupling character oscillates between ferromagnetic and antiferromagnetic order as a function of the thickness of spacer. The conduction electrons in the spacer layers mediate this effect. The RKKY model [6-8] is used for the periodic ion of the oscillation period of  $J_{12}$  for noble metal spacer [4], whose Fermi surface is fairly simple. In this model, the magnetic layers are described as arrays of localized spins interacting with conduction electrons by a contact exchange potential. Consequently, we might expect the  $T_c$  of a periodic stack of ferromagnetic layers to oscillate as a function of thickness nonmagnetic layers.

In the case that  $J_{12} = 0$ , we have isolated layers. In this case the magnetization depends sensitively on the symmetry of the order parameter. If the magnetization is kept to the plane by dipolar forces and then along one special direction in the film by anisotropy then we are in the Ising universality class and the Onsager [9] solution is appropriate. If the order parameter has continuous symmetry then the situation is more complex. We shall assume here that the isolated films do order as is observed in the experiment and hence an Ising approach is sensible. However, when  $J_{12} \neq 0$ , the Ising model becomes non-trivial and quite interesting both from a practical and theoretical point of view, with relation to the problem of two interacting fermionic fields. In this case an exact solution is lacking.

In the absence of an exact solution for three-dimensional ordering magnetic multilayers, some models are used for solving the problem. A number of attempts to go beyond the well-known statistical mechanical solution to the two-dimensional Ising problems have been proposed for systems of coupled two-dimensional Ising planes [0-13]. The system of two coupled, identical Ising planes ( $J_1 = J_2 = J_{12} = J_0$ ) was first studied by Ballentine [14] who investigated the model by high temperature series expansions in the case of  $J_{12} = J_0$ .

Ferronberg and Landau [15] considered the same two-layer problem using Monte Carlo simulations and the mean-field theory, then some other models are used [16-17].

All these theories give the ordering temperature of coupled planes to be higher than uncoupled planes ( $T_c > T_{uc}$ ). This does not agree with experiment [2]. We argue that the missing ingredient is an element of randomness introduced by fluctuation in the spacer layer thickness [18].

In multilayers magnetic thin films, we have interference between transmission and reflection spin due to the different path travelling from different layers (magnetic and nonmagnetic) like light in an optical Fabry-Perot cavity and thin films. Deviations from uniformity will affect the different magnetic properties in the different ways. Roughness in interface creates local thickness fluctuations. If there are variations in the spacer thickness of multilayers, due to polarization of free electrons in metallic spacer by the neighbouring magnetic planes, each plane will experience fluctuations in the inter-planar coupling. These effects can be induced in random fields (RF) on the magnetic planes.

For investigating Curie temperature of multilayers thin films, the two-dimensional Ising model with RF was chosen, because the strong easy axis, which gives order to an isolated plane, is in the plane for the thin magnetic layers of Ni ( $t_{Ni} \leq 4 ML$ ) [19]. The random-field Ising model [20] has been an important focus of theoretical and experimental studies of the statistics of random and frustrated systems [21]. This model is used in dilute antiferromagnet [22] and physics of spin glasses [23]. The simplest way to treat this problem is by the mean-field theory. This was carried out for a model with Gaussian and square distributions of RF's [18]. It was shown that the effect of a RF on a two-dimensional Ising ferromagnetic lies in the Curie temperature of ferromagnet and below the Curie temperature of the plane without randomness [24-26]. The approximation is an improvement over the mean-field theory which is the Bethe-Peierls-Wiess approximation (BPW), since it takes into account specific short-range order. The goal of this research is to investigate an expression for the paramagnetic susceptibility,  $\chi_p(T)$ , of an Ising plane in the presence of RF with square distributions (SD) in BPW when the RF's are varying very slowly over a correlation length. By using the mean field theory between the planes, the expression for the susceptibility of multilayers is used to obtain the  $T_c$  when the  $J_{12}$  is weak.

In the BPW, we first choose a "central spin",  $\sigma_0$ . The spin  $\sigma_0$  and its  $q$  nearest-neighbours  $\sigma_1, \sigma_2, \sigma_3, \sigma_4$  ( $q=4$ ) are treated exactly while the effect of the remainder of the lattice on the nearest-neighbours shell is taken into account in the mean-field approximation. In other words, we imagine that a cluster consisting of the "central spin"

and its  $q$  nearest-neighbours is immersed in the background provided by the remainder of the spins in such a way that each of the spins  $\sigma_1, \sigma_2, \sigma_3, \sigma_4$  experiences an effective local field,  $H$ , created by the spins outside this  $q+1$ -spin "cluster". We first evaluate the thermal average magnetization of the site centre of cluster,  $\langle \sigma_0 \rangle$ , and its neighbours,  $\langle \sigma_i \rangle$ , as in the normal BPW with RF, then equate  $\langle \sigma_0 \rangle$  to  $\langle \sigma_i \rangle$  to find the effective field,  $H$ , and then takes the average over the RF (the configuration average over the random fields). This corresponds to the solution of a problem in which the RF is varying smoothly over a correlation length.

In this approximation the Hamiltonian of the  $q+1$ -spin "cluster" is given by

$$H = -J_0 \sum_i^q \sigma_0 \sigma_i - (h + h_0) \sigma_0 - \sum_i^q (H + h_i) \sigma_i, \quad (1)$$

where the first term describes the Ising interaction between  $\sigma_0$  and its nearest neighbours,  $H$  is the uniform external magnetic field on the shell,  $h$  is the external field,  $h_0$  is RF acting on site 0,  $h_i$  is RF acting on neighbours that is different from  $h_0$  because we should look at the problem as two spins, one in the centre of cluster and one in the neighbourhood. In real problem  $h_i$  is the resultant of RF's of neighbours, so its amplitude is different from the site centre ( $h_0$ ). The partition function of the  $q+1$ -spin "cluster" is, therefore,

$$z = \sum_{\sigma_0, \sigma_i = \pm 1} \exp(-\beta H) = \sum_{\sigma_0, \sigma_i = \pm 1} \exp\left\{ J_0 \sum_i^q \sigma_0 \sigma_i + (h + h_0) \sigma_0 + \sum_i^q (H + h_i) \sigma_i \right\},$$

where  $\beta = 1/k_B T$ ,  $k_B$  is Boltzman constant, and  $T$  is temperature. For a plane with square lattice, the right hand side of this equation can be written as a sum of two terms

$$z = z_- + z_+,$$

where

$$z_{\pm} = \exp(\pm \beta(h + h_0)) \cosh^4(\beta(H \pm J_0 + h_i)).$$

Now, the thermal average of magnetization of the central spin is given by

$$\langle \sigma_0 \rangle = \frac{1}{\beta z} \frac{\partial z}{\partial h} = \frac{z_+ - z_-}{z_+ + z_-}. \quad (2)$$

Similarly, the thermal average of magnetization on sites neighbouring can be calculated from

$$\langle \sigma_i \rangle = \frac{1}{q} \sum_i^q \sigma_i = \frac{1}{q\beta z} \frac{\partial z}{\partial H},$$

and, hence

$$\langle \sigma_i \rangle = \frac{1}{z_+ + z_-} \quad (3)$$

$$(z_+ \tanh \beta(J_0 + H + h_i) + z_- \tanh \beta(-J_0 + H + h_i)).$$

Since the ferromagnet is translationally invariant, we must have  $\langle\sigma_0\rangle=\langle\sigma_1\rangle$ . This leads to the self-consistency condition that  $H$  is determined by

$$2\beta(-h+H-h_0+h_1) = 3Ln[\cosh\beta(J_0+H+h_1)\operatorname{sech}\beta(-J_0+H+h_1)]. \quad (4)$$

Now we have to solve the eq. (4) for the unknown effective field  $H$ . By using the equation

$$\tanh x \tanh y = \tanh y(1/2 \ln(\cosh(x+y)\operatorname{sech}(x-y))),$$

in the eq.(4), the effective field is obtained as

$$H = h + \frac{3 \arctanh(\tanh J_0 \beta \tanh \beta(H+h_1))}{\beta} + h_0 - h_1. \quad (5)$$

Then, the thermal average of magnetization is given as

$$\langle\sigma\rangle = \tanh(4 \arctanh(\tanh J_0 \beta \tanh \beta(H+h_1)) + \beta(h+h_0)). \quad (6)$$

By replacing eq. (5) in eq.(6), we can write the expression of the thermal average magnetisation as

$$\langle\sigma\rangle = \tanh(1/3 \beta(h-4H-4h_1+h_0)). \quad (7)$$

This equation is a function of RF's.

For any symmetric distribution of RF's we can write

$$\int_{-\infty}^{+\infty} p(h_i) dh_i = 1.$$

The configuration average over the RF's  $\{h_i\}$  for every site is given as

$$\langle\langle\alpha\rangle\rangle = \int_{-\infty}^{+\infty} \langle\alpha_i\rangle p(h_i) dh_i, \quad (8)$$

where  $p(h_i)$  is the distribution function, and  $\langle\alpha_i\rangle$  is the thermal average of magnetization. The probability for RF with square distribution is given as

$$p(h_i) = \begin{cases} 1 & \text{if } |h_i| \leq \Delta \\ 0 & \text{if } |h_i| \geq \Delta \end{cases},$$

then the average of the magnetization,  $\langle\langle\sigma\rangle\rangle$ , over RF's  $h_0$  and  $h_1$  is given by

$$\langle\langle\sigma\rangle\rangle = \frac{1}{2\Delta} \int_{-\Delta}^{+\Delta} \frac{1}{2\delta} \int_{-\delta}^{\delta} dh_0 dh_1 p(h_0) p(h_1) \langle\sigma\rangle = \frac{1}{4\delta \Delta} \int_{-\Delta}^{+\Delta} \int_{-\delta}^{\delta} dh_0 dh_1 \tanh^{-1}(1/3 \beta(h-4H-4h_1+h_0)), \quad (9)$$

where  $2\delta$  and  $2\Delta$  are full-width of  $h_0$  and  $h_1$ , respectively. The double integrals of the  $\langle\langle\sigma\rangle\rangle$  is not so difficult to evaluate with the square distribution. The susceptibility of a plane in BPW is given as

$$\chi_p = d\langle\langle\sigma\rangle\rangle/dh = \partial\langle\langle\sigma\rangle\rangle/\partial h + (\partial\langle\langle\sigma\rangle\rangle/\partial H)(dH/dh). \quad (10)$$

We follow the same procedure as was done on  $\langle\langle\sigma\rangle\rangle$  for  $\langle dH/dh \rangle$ , this means that one integrates the expression

$dh/dH$  over the  $h_0$  and  $h_1$  with square distributions. By using the expressions  $\partial\langle\langle\sigma\rangle\rangle/\partial h$ ,  $\partial\langle\langle\sigma\rangle\rangle/\partial H$  and  $\langle dH/dh \rangle$  in eq. (10),  $\chi_p$  is given as

$$\chi_p = \frac{3}{16\beta\delta\Delta} \left( 1 - \frac{16\beta\Delta}{4\beta\Delta + 3Ln\left(\frac{\cosh(\beta(H+J_0-\Delta))\cosh(\beta(H-J_0+\Delta))}{\cosh(\beta(-H+J_0+\Delta))\cosh(\beta(H+J_0+\Delta))}\right)} \right) \left( Ln\left(\frac{\cosh(\beta/3(h-4H+\delta-4\Delta))\cosh(\beta/3(-h+4H+\delta-4\Delta))}{\cosh(\beta/3(h-4H+\delta+4\Delta))\cosh(\beta/3(-h+4H+\delta+4\Delta))}\right) \right).$$

At high temperature ( $T_c \geq T$ ), when  $H \approx 0$ ,  $h \approx 0$ , and by defining  $\Delta = \varepsilon\delta$  where  $\varepsilon$  is a constant, the  $\chi_p$  is given by

$$\chi_p = \frac{3}{8\varepsilon\beta\delta^2} \left( 1 - \frac{8\varepsilon\beta\delta}{2\beta\varepsilon\delta + 3Ln\left(\frac{\cosh(\beta(J_0-\varepsilon\delta))}{\cosh(\beta(J_0+\varepsilon\delta))}\right)} \right) Ln\left(\frac{\cosh(\beta/3(\delta-4\varepsilon\delta))}{\cosh(\beta/3(\delta+4\varepsilon\delta))}\right). \quad (11)$$

When  $\delta \rightarrow 0$  (zero randomness), the eq (11) simplify as

$$\chi_p = \frac{\beta(1 + \tanh J_0 \beta)}{1 - 3 \tanh J_0 \beta}. \quad (12)$$

We recover the usual  $\chi_p$  of BPW.

The inverse susceptibility,  $1/(J_0\chi_p)$ , of square and Gaussian distribution (from papers [27]) were plotted (figure 1) when  $\varepsilon=1$  and  $\delta/J_0=0.3$ . The plot shows that  $1/(J_0\chi_p)$  is zero at smaller temperature when the RF is present. It confirms a divergence at a reduced value of  $T_c$ . The reduction of  $T_c$  is smaller for long correlation length, and bigger for Gaussian than square distribution. Ordering temperature of coupled planes,  $T_c$ , is given by the solution of the equation  $2|J_{12}|\chi_p=1$  that in the previous paper was obtained [18]. In this model,  $T_c$  depends on  $|J_{12}|$ ,  $\delta$  and  $\varepsilon$ . and can be higher or lower than the Curie temperature of an isolated plane.

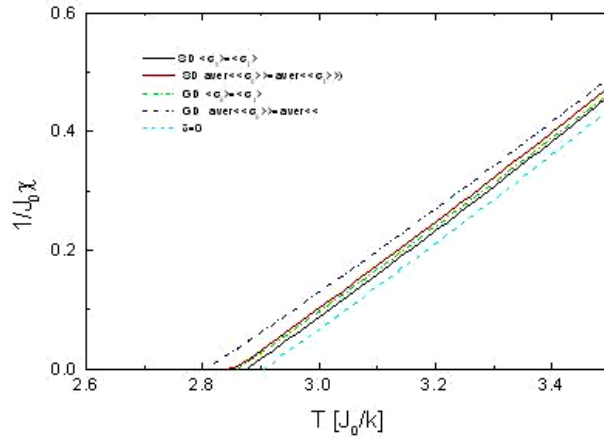
We present the theoretical results on the  $T_c$  of well characterized Ni/Au/Ni multilayers which was measured by Bayreuther, et.al., [2]. In a multilayer  $J_{12}$  is an oscillating function [1] of the spacer thickness,  $t$ . Suppose  $J_{12}$  is given by

$$J_{12} = Ae^{-\gamma t} \cos(\Lambda t + \varphi). \quad (13)$$

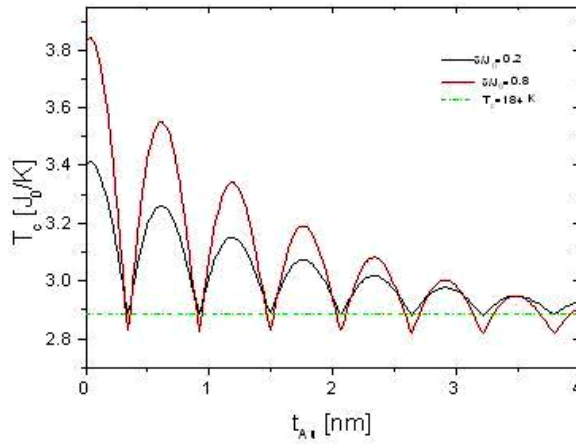
The halfwidth of the RF is given by

$$\delta = \left| \frac{\partial J_{12}}{\partial t} dt \right| \approx AAe^{-\gamma t} |\sin(\Lambda t + \varphi) dt|, \quad (14)$$

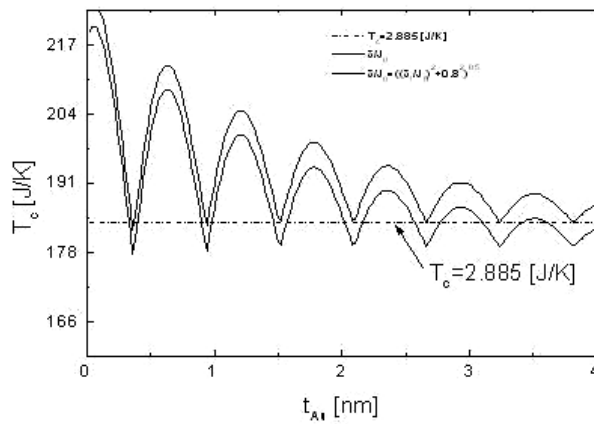
in which  $A$  is a constant,  $t$  is the thickness of spacer Au[111],  $\Lambda$  is the oscillation period. The observed oscillation period of the  $J_{12}$  for noble metal spacers is  $A_{exp}=(1.15\pm 0.1) \text{ nm}$  [2], which is in good agreement with the theoretical value ( $A_{th}=1.135\text{nm}$ ) [4]. The parameters of  $J_{12}$  are calculated from the paper [2]. In this calculation, the theoretical  $J_{12}$  in magnetic surface, Ni, at



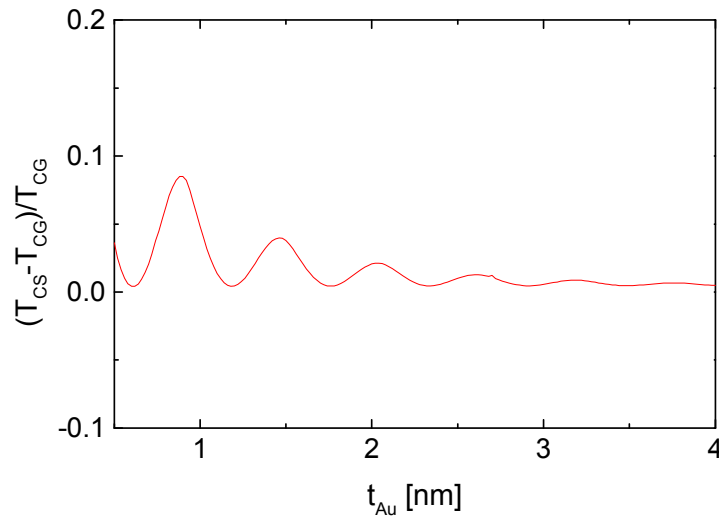
**Figure 1.**  $1/J_0\chi_p$  plotted as a function of temperature with square and Gaussian distribution when  $\text{aver}\langle\sigma_i\rangle=\text{aver}\langle\sigma_j\rangle$  then obtained  $H$  [18] and when  $\langle\sigma_i\rangle=\langle\sigma_j\rangle$  then take average over the RF's, next obtained the  $H$ . ( $\delta J_0=0.3$  and  $\Delta=\delta$ ).



**Figure 2.**  $T_{cs}$  plotted as a function of  $t_{Au}$  for different strengths of RF's ( $\delta J_0=0.2$  ,  $\delta J_0=0.8$ ) in BPW model. The dash-dot line is for Curie temperature of isolated plane.



**Figure 3:**  $T_{cs}$  plotted as a function of  $t_{Au}$  ( $\Delta=\delta$ ,  $dt=0.5\text{nm}$ ), random fields is defined as  $\delta=(\delta_1^2+\delta_2^2)^{1/2}$ , that  $\delta_1=|(\partial J_{12}/\partial t)|$  ( $\delta_2/J_0=0.0$ ,  $\delta_2/J_0=0.8$ ) in BPW model



**Figure 4.**  $(T_{cs}-T_{cg})/T_{cg}$  plotted as a function of thickness of spacer,  $t_{Au}$  when  $\langle\sigma_0\rangle=\langle\sigma_1\rangle$ .

first antiferromagnetic is  $0.4 \text{ erg/cm}^2$  [4] and experimentally from the effective spin wave parameter is around  $0.5 \text{ erg/cm}^2$  [2], then  $A=0.2412$ ,  $\varphi=-0.30351$ ,  $A=5.46364 \text{ rad/nm}$  and  $\gamma=0.61151/\text{nm}$ .

For a magnetic layer with thickness  $t$ , the  $T_c$  of an magnetic layer depends on the thickness of the layer [28-29]. The  $T_c$  of an uncoupled plane for  $t_{Ni}=0.73 \text{ nm}$  is  $184 \text{ K}$  [2]. RF is assumed to arise because of fluctuations in  $J_{12}$  due to variations in the spacer thickness. It is better to consider the RF as being related to the value of the  $J_{12}$ . Since  $J_{12}(t)$  is an oscillatory function of  $t$  its derivative  $dJ_{12}/dt$  is zero just when  $J_{12}(t)$  passes through one of its maxima or minima, and a maximum as  $J_{12}(t)$  passes through zero. Hence a small variation in the spacer thickness  $\delta t$  will give a vanishingly small effect when  $|J_{12}(t)|$  is large but a larger effect where  $J_{12}(t) \rightarrow 0$ . Hence where  $J_{12}(t) \rightarrow 0$  the  $T_c$  is given by the divergence of  $\chi_p$  in the presence of RF given in terms of the spacer fluctuations,  $\delta t$ .

$T_c$  is shown as a function of thickness of Au,  $t_{Au}$  in figures 2, when RF's are constant ( $\delta/J_0=0.0$ ,  $\delta/J_0=0.8$ ), this plot shows that the RF reduces the ordering temperature of coupled planes, the more the strength of RF the more the reduction of  $T_c$ .

When RF's vary as  $\delta=(\delta_1^2+\delta_2^2)^{1/2}$ , where  $\delta_j=(\partial J_{12}/\partial t)dt$  with  $dt=0.5 \text{ nm}$  and constant RF,  $\delta_2/J_0=0.8$ . In some parts of this plot, the  $T_c$  is less than the  $T_c$  of the uncoupled plane (dashed line) because of the RF and in some parts it is greater because of the  $J_{12}$  and it oscillates with half period of  $J_{12}$  (figure 3). Moreover, the depths of valleys are different and are in agreement with the experiment.

In figure 4, the relative variance of ordering temperature is plotted as a function of spacer thickness. This plot shows that the ordering temperature of multilayer depends on the distribution of RF at the small thickness of spacer.

Finally, we have analyzed the RF Ising model in the BPW with the square distribution with long correlation length and extend the analysis of the previous paper [18]. It is shown that the  $\chi_p$  of a plane with Gaussian distribution of RF decreases more than the square distribution. When the correlation length is long ( $\langle\sigma_0\rangle=\langle\sigma_1\rangle$ ), the decreasing in  $T_c$  in the square is less than Gaussian distribution. Our results show that the  $T_c$  changes as a half period of  $J_{12}$ . Without randomness all ordering temperatures of coupled planes are more than the uncoupled planes. This does not agree with the experimental results. By increasing the constant strength of RF, all  $T_c$ 's go down relative to the uncoupled plane. In the cases  $\delta$  vary as  $\delta=(\delta_1^2+\delta_2^2)^{1/2}$ , where  $\delta_j=(\partial J_{12}/\partial t)dt$  and  $\delta_2/J_0=\text{const}$ , in some parts of the thickness of Au, the  $T_c$  is less than the  $T_c$  of uncoupled plane (dash line) and in some parts more than it. In experimental results, the depths of valleys are different ???. These results show that  $T_c$  depends on the distributions of RF at small thickness of the spacer.

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